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Electronic spin susceptibilities and superconductivity in $HgBa_2CuO_{4+\delta}$ from nuclear magnetic resonance

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Nuclear magnetic resonance (NMR) experiments on single crystals of $HgBa_2CuO_{4+\delta}$ are presented that identify two distinct temperature-dependent spin susceptibilities: one is due to a spin component that is temperature-dependent above the critical temperature for superconductivity (T_c) and reflects pseudogap behavior; the other is Fermi-liquid-like in that it is temperature independent above T_c and vanishes rapidly below T_c. In addition, we demonstrate the existence of a third, hitherto undetected spin susceptibility: it is temperature independent at higher temperatures, vanishes at lower temperatures (below $T_0 \neq T_c$), and changes sign near optimal doping. This susceptibility either arises from the coupling between the two spin components, or it could be given by a distinct third spin component. Recent susceptibility data on single crystals support its presence in most cuprates.

The high-temperature superconducting cuprates are still in the focus of condensed matter physics, and while their properties are rather complex, they give rise, e.g., to a more or less unique, simple dependence of NMR shifts on temperature and doping, caused by the uniform magnetic susceptibility. The data in Fig. 1 are rather typical and can serve as a good example: at high doping levels *and* high temperatures, the shifts are rather independent of temperature, and they rapidly decrease below T_c (reminiscent of a Fermi liquid with spin singlet pairing). As the doping level is lowered, the pseudogap makes the shifts temperature-dependent even above T_c, whereas the sudden decrease below T_c disappears.

A long-standing question has been whether a single electronic fluid's temperature-dependent electronic spin polarization, $S(T) = \chi(T)B_0$, in a magnetic field (B_0) can explain these shifts. From the analyses of $YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_4O_8$ shifts measured at planar copper and oxygen above and below T_c it was concluded that this is the case[1,2]. NMR shift experiments measure $\chi(T)$ rather reliably since magnetism due to impurities is typically not problematic and NMR can even access shifts below T_c. $\chi(T)$ must cause proportional spin shifts at *all* nuclei, and for a given orientation (η) of a crystal with respect to B_0 we expect a spin shift $K_{S\eta}(T) = q_\eta \cdot \chi(T)$, where the anisotropy of the shift is carried by the effective hyperfine coefficients (q_η).

So far, NMR shifts have been interpreted with an isotropic $\chi(T)$, but even an anisotropic susceptibility could be described with a different q_η. In fact, for the proof of single component physics[1,2], the copper shifts were measured with B_0 in the CuO_2 plane, while for oxygen B_0 was perpendicular to it (the shift in the other directions are too small or not well defined for powder samples). While there were doubts from susceptibility measurements about the single component view early on[3,4], the isotropic response was questioned only more recently[5,6].

A few years ago, it was shown with NMR that the spin shifts at Cu and O in $La_{1.85}Sr_{0.15}CuO_4$ cannot be explained with a single spin component's $\chi(T)$, but rather require two spin components with distinctly different temperature dependencies[7]. One of the components, $S_1(T)$, causes the pseudogap response, and it dominates the planar O shift. The second component, $S_2(T)$, is temperature independent above T_c (Fermi-liquid-like) and rapidly vanishes below it. The second component dominates the planar Cu and apical O shifts. Since a possible anisotropy in $S_{1,2}$ would be contained in the hyperfine couplings we proceed with isotropic $S_{1,2}$ and discuss consequences from their possible anisotropy later. Then, S_1 and S_2 affect a nucleus through $q_{1\eta}$ and $q_{2\eta}$, respectively, so that its spin shift is

$$K_{S\eta}(x,T) = q_{1\eta} \cdot \chi_1(x,T) + q_{2\eta} \cdot \chi_2(x,T). \qquad (1)$$

We note that if S_1 and S_2 are coupled, χ_1 and χ_2 must be the sum of two terms each, i.e., $\chi_1 = \chi_{11} + \chi_{12}$ and $\chi_2 = \chi_{12} + \chi_{22}$, where χ_{12} is the coupling susceptibility that describes how S_1 responds to a magnetic field acting on S_2[7–9].

Motivated by the results for $La_{1.85}Sr_{0.15}CuO_4$, we investigated another single-layer system, $HgBa_2CuO_{4+\delta}$. With ^{63}Cu and ^{199}Hg NMR on underdoped (T_c=74 K, UN74) and optimally doped (T_c=97 K, OP97) single crystals, the failure of a single component approach became apparent as well[9]. However, the doping dependence of the temperature independent component remained unclear[9]. The reason for this will be uncovered here as we identify a new shift component: it is temperature independent at high temperatures and vanishes at low temperatures, but it differs from the Fermi-liquid-like component in that it changes sign as a function of doping (it is nearly zero for optimal doping). Furthermore,

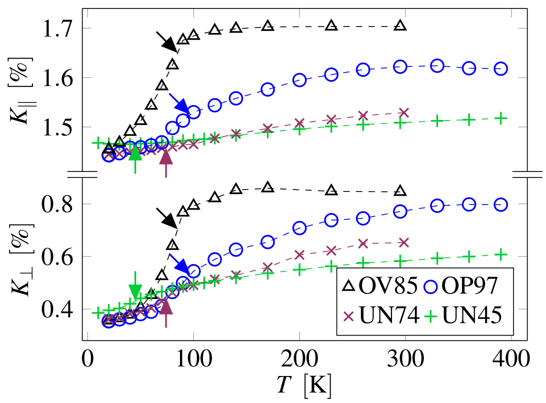

FIG. 1. (Color online) Total magnetic ^{63}Cu shifts K_η as a function of temperature. Upper panel: B_0 parallel to the crystal c-axis ($K_\parallel$); lower panel: B_0 in the CuO_2 plane ($K_\perp$). For $K_\perp$, the contribution from the quadrupole interaction was removed. Dashed lines are guides to the eye. Arrows indicate T_c values. Errors are smaller than the data point size.

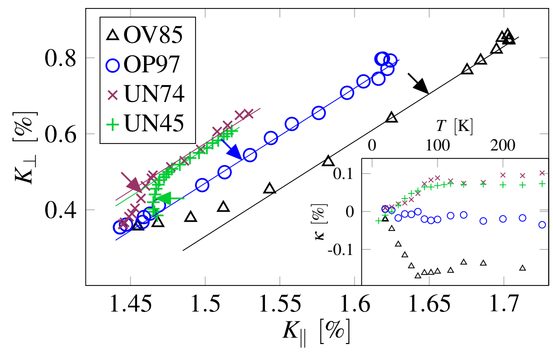

FIG. 2. (Color online) $K_\perp(T)$ versus $K_\parallel(T)$ with temperature as an implicit parameter. Arrows indicate T_c values. The straight lines have the slope 2.5 obtained from the fit to the data down to T_0. Inset shows $K_{S,\perp}(T) - 2.5K_{S,\parallel}(T)$ as a function of temperature.

	x	T_0		x	T_0
UN45	0.06(1)	80(10) K	OP97	0.16(2)	≈75(10) K
UN74	0.10(1)	80(10) K	OV85	0.19(1)	60(10) K

TABLE I. Values of doping level x[11] and T_0.

the characteristic temperature (T_0) at which it suddenly begins to disappear, depends only weakly on doping and can be larger than T_c for underdoped, and lower than T_c for overdoped samples. Since T_0 is similar to T_c for UN74 this component was not identified earlier[9]. We argue below that this new component is likely a generic property of all cuprates and is connected to the newly identified anisotropic susceptibility[6].

Two new $HgBa_2CuO_{4+\delta}$ single crystals with T_c=45 K (UN45) and 85 K (OV85) were prepared following the method described previously[10,11]. The experimental details of exciting, recording and referencing the ^{63}Cu NMR signals are identical to those in Ref.[9,12,13]. It was also shown that the diamagnetic response due to the mixed state below T_c can be neglected for ^{63}Cu shifts, making them very reliable also below T_c[9].

In Fig. 1, we show the measured ^{63}Cu shifts, $K_\parallel(T)$ and $K_\perp(T)$, for all $HgBa_2CuO_{4+\delta}$ single crystals studied (including those from Ref.[9]). We display the total experimentally measured magnetic shift, $K_\eta(T) = K_{L\eta} + K_{S\eta}(T)$, which is the sum of a temperature and doping *independent* orbital part ($K_{L\eta}$)[14] and the temperature and doping *dependent* spin part ($K_{S\eta}$).

In Fig. 2, we show the same data, but plotted as $K_\perp(T)$ versus $K_\parallel(T)$. At higher temperatures (large shift values) we observe parallel lines that begin to approach a common low-temperature point below a characteristic temperatures $T_0 \neq T_c$ (cf. Tab. I). This implies the presence of a shift component that is temperature-independent at high temperatures, but disappears below T_0. With just the data for UN74 and OP97 it was erroneously concluded[9] that this offset between the parallel lines is due to the Fermi-liquid-like component. In order to analyze the data in Fig. 2 we write,

$$K_{S\perp}(T) = \frac{1}{c_0} K_{S\parallel}(T) + \kappa(x,T), \quad (2)$$

where $\kappa(x,T)$ describes the temperature dependent offset between the parallel lines, which is plotted in the inset in Fig. 2. We adopt the typical definition of the spin shift ($K_{S\eta}$) by choosing $K_{L\eta}$ as the remaining shift at the lowest temperatures, i.e., $K_{S\eta}(T) = K_\eta(T) - K_{L\eta}$, but the basic findings do not depend on the choice of $K_{L\eta}$. For the common high temperature slope we determine $c_0 \approx 0.40 \pm 0.02$. We are rather certain that the new shift component is due to a spin susceptibility (χ_κ), i.e., $\kappa(x,T) \propto \chi_\kappa$. While it is a natural assumption, we find evidence for $\kappa(x,T)$ also in ^{199}Hg NMR[9], and recent ^{17}O NMR[15] (see Supplement), as well as in recent susceptibility measurements[6].

We can learn more about the shift components just from the highly reliable Cu shifts. As reported earlier[9,12], the pseudogap shift component ($K_{S,PG}$) has a unique temperature dependence, at least up to optimal doping: $K_{S,PG}(x,T) = x \cdot \sigma(T)$, where x is the average doping level of the sample and $\sigma(T)$ a universal function of temperature. Our new data support this scaling, and we explain in more detail in the Supplement that it is in quantitative agreement with susceptibility data[3,4,6] for the pseudogap susceptibilities of other cuprates and ^{17}O shifts in $Bi_2Sr_2CaCu_2O_{8+\delta}$[16]. As a consequence, if one plots the shifts measured on samples with different doping levels against each other (with temperature as an implicit parameter), straight lines or line segments are found. This can be seen in Fig. 3, and indeed, the slopes of the linear segments are equal to the doping ratios. (A similar scaling was also observed for the electronic entropy of $YBa_2Cu_3O_{6+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$[17]).

We now discuss Fig. 3 in more detail to motivate the ensuing numerical analysis. First, we consider UN45 and

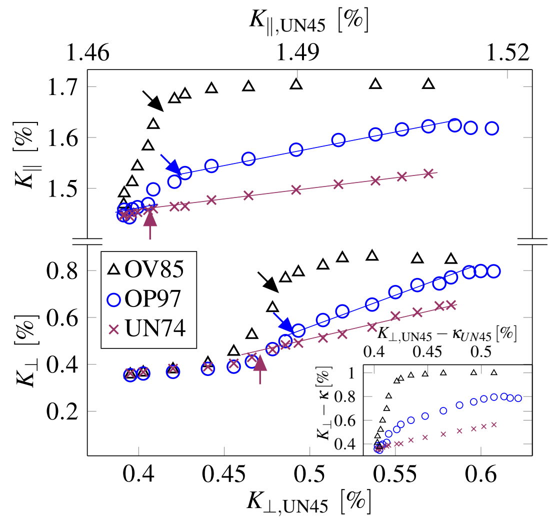

FIG. 3. (Color online) $K_\|$ (upper panel) and $K_\perp$ (lower panel) of the UN74, OP97 and OV85 samples plotted versus shifts of the UN45 sample with temperature as an implicit parameter. Straight lines have slopes derived from doping ratios. Inset shows $K_\perp - \kappa$ of UN74, OP97 and OV85 samples versus $K_\perp - \kappa$ of the UN45 sample.

UN74. For $c \parallel B_0$, the shifts for these two samples are nearly proportional to each other (in the whole temperature range), also for $c \perp B_0$ after subtracting $\kappa(x,T)$ (cf. inset in Fig. 3). With the proportionality of the two shifts, not interrupted near either sample's T_c, we conclude that any shift due to S_2 must be negligible.

Next, we examine OP97 (for which $\kappa \approx 0$, cf. Fig. 2). As concluded earlier[9], in a broad temperature range above and below T_c we find the expected slope for both orientations (Fig. 3). The sudden change of $K_{\|,\mathrm{OP97}}$ near 97 K must then be due to S_2. This means that the anisotropies due to both spin components, S_1 and S_2, are the same, so that the corresponding changes in the shifts do not show any discontinuities in Fig. 2.

We now turn to OV85. Going back to Fig. 1, we notice that $K_\eta(T)$ is nearly constant above T_c, but starts to rapidly decrease at T_c (as if dominated by S_2). Fig. 2 reveals that this decrease begins well above the temperature T_0 below which $\kappa(T)$ begins to change (i.e., when the slope in Fig. 2 changes). Again, this says that the two shift components due to S_1 and S_2 share the same anisotropy.

To conclude, we have identified three spin shift components that differ in their temperature and doping dependence, and since two of them share the same anisotropy we analyze all shifts numerically with,

$$K_{S\eta}(x,T) = q_{1\eta}\left[\chi_1(x,T) + \chi_2(x,T)\right] + q_{\kappa\eta}\chi_\kappa(x,T). \quad (3)$$

We make the following assumptions: (1), the pseudogap shifts obey the scaling behavior ($K_{S,PG}(x,T) = x \cdot \sigma(T)$); (2), for UN45 and UN74 the shifts are only given by $q_{1\eta}\chi_1$ and $q_{\kappa\eta}\chi_\kappa$ since there are no shift changes at T_c; (3), $q_1\chi_2$ is constant above T_c; and, (4), the shift

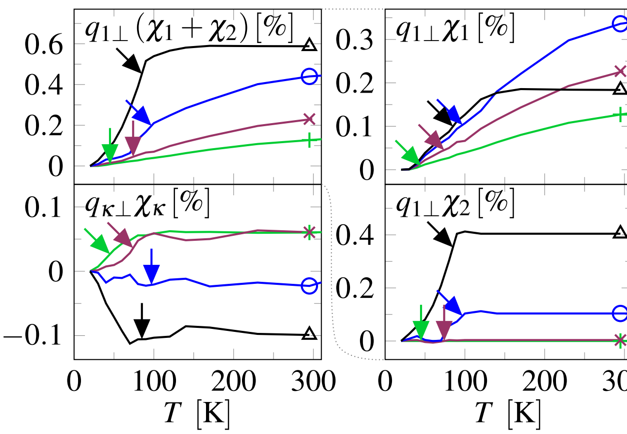

FIG. 4. (Color online) Results of the numerical decomposition of the spin shifts for $c \perp B_0$. Left panel: into $q_{1\perp}(\chi_1 + \chi_2)$ and $q_{\kappa\perp}\chi_\kappa$ according to (3). Right panel: into the pseudogap component $q_{1\perp}\chi_1$ and the Fermi-liquid-like component $q_{1\perp}\chi_2$. The arrows indicate T_c values, symbols are only to help identify the samples.

components for $c \perp B_0$ and $c \parallel B_0$ are proportional (i.e., the anisotropy of the shift can originate either from the hyperfine coefficient or the susceptibility). This leads to the results displayed in Fig. 4 for $c \perp B_0$ (the results for $c \parallel B_0$ differ only in magnitude due to anisotropy of $q_{1\eta}$ and $q_{\kappa\eta}$). A detailed description of the analysis is given in the Supplement.

The left panel of Fig. 4 shows the first step of the shift decomposition: we see how $q_{1\perp}(\chi_1+\chi_2)$ and $q_{\kappa\perp}\chi_\kappa$ evolve with temperature and doping. χ_κ changes sign near optimal doping and is almost twice larger in magnitude for OV85 than for the two underdoped samples. In the right panel of Fig. 4 we extract $q_{1\perp}\chi_1(T)$ and $q_{1\perp}\chi_2(T)$ using the scaling of χ_1. At low doping, $q_{1\perp}\chi_2$ is negligible, but rapidly increases with doping. For the temperature range of our study, χ_1 grows with increasing doping up to optimal doping. It can be identified even for OV85 at lower temperatures, but its high-temperature behavior cannot be reliably extracted (recently the presence of the pseudogap in overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ was confirmed by ARPES[18]).

Now, in view of recent susceptibility data[6] the question arises of what NMR can say about possible anisotropic spin susceptibilities that have been assumed to be negligible for the interpretation of the shifts in cuprates.

The early work concerning single-fluid behavior[2,19] concentrated on $YBa_2Cu_3O_{6.63}$, and $YBa_2Cu_4O_8$, both are underdoped cuprates. In both cases, the planar Cu shifts for $c \perp B_0$ were found to have the same temperature dependence as the planar O shifts for $c \parallel B_0$. Similarly, a unique temperature dependence of all shifts (including apical oxygen) was reported in $La_{1.85}Sr_{0.15}CuO_4$ above about T_c[7]. These findings, together with the scaling behavior of χ_1 discussed above, which holds throughout most of the phase diagram even in the presence of other shift components, very likely mean that $\chi_1(T)$ must have a temperature independent or vanishing anisotropy.

$\chi_2(T)$, the Fermi-liquid-like component, is temperature independent above T_c. While it could have a temperature dependent anisotropy below T_c, it appears unlikely since the overall shift change below T_c is in agreement with the scenario above T_c.

Therefore, our newly discovered third component is of particular interest. Fig. 2 shows parallel lines above T_0 even for samples with $T_c > T_0$, which demands the same anisotropy for shift changes due to $\chi_{1,2}$. The fixed offset between the lines is set by a doping dependent χ_κ, which could have an anisotropy that becomes even temperature-dependent below T_0.

We would like to point out that κ cannot be explained by a redistribution of NMR spectral weight with temperature within the rather broad Cu resonance. This is also seen from the Hg NMR linewidths[9], since they are smaller than the changes due to κ.

If χ_1 is the susceptibility of S_1 and χ_2 that of S_2, χ_κ could be due to the coupling between S_1 and S_2, i.e., $q_{\kappa\eta}\chi_\kappa(x,T) = 2q_{1\eta}\chi_{12}$. As such, the sign change of χ_κ with doping may indicate a change in sign of the electronic spin-spin coupling. Since the apparent anisotropies of $q_{1\eta}$ and $q_{\kappa\eta}$ are different, χ_κ would have to be anisotropic. Alternatively, if χ_κ were the susceptibility of a new spin component (S_3) coupling of S_3 to S_1 and S_2 could possibly be leading to a complicated shift scenario that can, however, be described in a rather simple way as shown here.

The fact that the Cu nucleus sees the same anisotropies for S_1 and S_2 is perhaps not surprising, but argues against a trivial picture of different Cu and O spins to which a Cu nucleus would couple with different angular dependencies (the anisotropies of such spins could be different, as well). Perhaps, S_1 and S_2 relate to antinodal and nodal quasi-particles, respectively, which may be coupled to give χ_κ[20], but we did not attempt to use a particular model to separate possible contributions[6,21]. For example Pines and Barzykin explained the temperature and doping dependence of the uniform spin susceptibility of $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$, assuming coexistence of two electronic fluids: a two-dimensional local moment spin liquid and a quasiparticle fermion liquid[22,23].

Interestingly, new susceptibility measurements on cuprate single crystals appear to converge on the finding of an anisotropic uniform susceptibility, i.e., $\chi_\parallel(T) = 1.4[\chi_\perp(T) - I(x)]$ above T_c, where $I(x)$ depends on doping[6]. This relation is of similar form as (2), and suggests that our anisotropic shift is indeed caused by an anisotropic susceptibility. However, our measurements extend to lower temperatures where we find this anisotropic shift (and susceptibility) to disappear.

The scenario found here reminds one of a quantum critical point near optimal doping[24] (where χ_κ changes sign): on the underdoped side we have χ_1 and χ_κ, on the overdoped side χ_2 and χ_κ. It is not clear whether the Fermi-liquid-like behavior in the underdoped region observed in other experiments (d.c. resistivity, optical conductivity, and magnetoresistance measurements) on $HgBa_2CuO_{4+\delta}$[25–27] corresponds to a small Fermi-liquid-like component (invisible to NMR) or is related to χ_κ. An important question to be addressed in future experiments is whether χ_κ and χ_2 are perhaps connected with the normal-state charge-density-wave correlations and the quantum oscillations observed below optimal doping[28–30].

To conclude, based on a detailed study of the local magnetic response of $HgBa_2CuO_{4+\delta}$ single crystals we confirm that a description of the NMR shifts with a single, temperature-dependent spin component is not possible. One shift component is due to the pseudogap and it governs the NMR shifts at lower doping levels. The second component shows Fermi-liquid-like behavior and governs on the overdoped side of the phase diagram, where the pseudogap shift is suppressed. We discovered a new, third shift component that could not be distinguished from the Fermi-liquid-like component, earlier[9]. The new component is temperature independent above a critical temperature T_0, which can be significantly larger than T_c for underdoped or smaller than T_c for overdoped crystals. Since it changes sign (near optimal doping), and it disappears below T_0 rather than T_c, it is very different from the Fermi-liquid-like component. From recent reports on the uniform susceptibility we conclude that this component must be present in all cuprates and is caused by an anisotropic spin polarization. Given its properties and the fact that on can rarely investigate the anisotropy of the NMR spin shift for a given nucleus with high precision, this third component could have easily been missed. With the evidence discussed for the various cuprates, and the recent proof that the closing of the pseudogap for one of the cornerstone single-component materials ($YBa_2Cu_4O_8$) with pressure readily reveals two-component behavior[31], there can be no doubt anymore that a multi-component analysis of the NMR data is necessary.

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